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New Adsorption Cycles for Carbon Dioxide Capture and Concentration

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Background

- generally accepted that increasing global temperatures over recent decades due to increasing atmospheric concentrations of greenhouse gases, i.e., CH_4, N_2O, and particularly CO_2
- carbon sequestration probably newest means being studied to manage CO_2 in the environment
- most likely options for CO_2 sequestration
  - chemical and physical absorption
  - low-temperature distillation
  - gas separation membranes and
  - physical and chemical adsorption

Objectives

- study new pressure swing adsorption (PSA) cycles for CO_2 capture and concentration at high temperature
  - two key features of these new PSA cycles
    - heavy reflux (HR) PSA concept
    - use of a hydrotalcite like (HTlc) adsorbent that captures CO_2 reversibly at high temperatures simply by changing pressure
  - bench-scale experimental and theoretical program being carried out to complement and extend the process simulation study that was carried out during Phase I (DE-FG26-03NT41799)
  - nine tasks being carried out over 3-year period

Tasks

Task 1. Construct Fixed Bed Unit (High Temperature System)
Task 2. Modify Existing PSA System to 4-Beds (Low Temperature System)
Task 3. Perform Experiments in the Fixed Bed Unit
Task 4. Perform Experiments in the 4-Bed PSA Unit
Task 5. Modify and Validate Existing PSA Code
Task 6. Carry Out Rapid Adsorbent Characterization
Task 7. Carry out Simulations with Validated PSA Code
Task 8. Carry out Detailed Adsorbent Characterization
Task 9. Carry out Economic Analyses

Significant Accomplishments

Publications
- 6 in print
- 4 submitted
- 4 in preparation

Presentations
- 16

Book Chapters
- 1

Invention Disclosures
- 1

PhD Students Produced
- 3

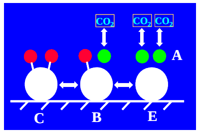
New Projects
- 4

CO_2 Uptake and Release from K-Promoted HTlc

How does CO_2 interact with K-promoted HTlc?

- recent studies describe adsorption and desorption behavior of CO_2 using simple Langmuir and linear driving force models
- none of them provide detailed mechanism of CO_2 equilibrium and kinetic behavior
- mechanistic understanding and realistic model needed for PSA process design
- proposed mechanism that describes CO_2 dynamics of uptake and release via reversible non-equilibrium kinetic model

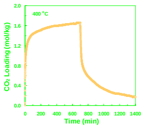
Non-Equilibrium Reversible Kinetic Model



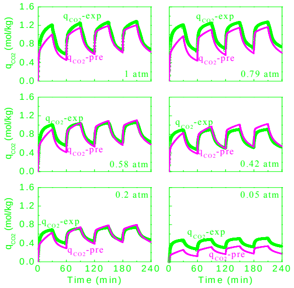
$A \equiv CO_2(ad)$
$E \equiv Mg_6Al_2K_2O_{10}$
$B \equiv Mg_6Al_2K_2O_9(CO_3)$
$C \equiv Mg_6Al_2K_2O_8(CO_3)_2$

Activation | Desorption | Adsorption

$I \rightarrow C + (n+8)H_2O$
$C \leftrightarrow B + A$ (slow)
$B \leftrightarrow A + E$ (intermediate)
$A \leftrightarrow CO_2(g)$ (fast)



T&P Dependent Model vs. Experiment at 400 °C

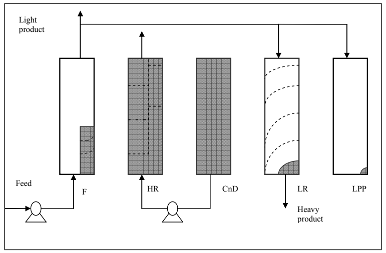


Equilibrium Theory Model of Heavy Reflux PSA

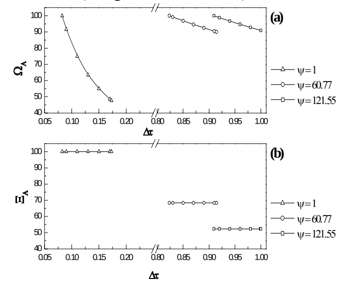
Why is an equilibrium theory model important?

- isothermal model with no non-ideal effects predicts the ultimate performance possible
- many analyses in the literature; none describe HR PSA cycles suitable for CO_2 capture
- information from such models indispensible to HR PSA Cycle understanding and limitations
- carried out fundamental analyses of HR PSA cycles, exposing interesting extreme cases

Schematic of Shock Wave and Simple Wave Development During Typical HR PSA Cycle



Variation of Heavy Component Recovery (Ω_A) and Purity (Ξ_A) with $\Delta\tau$ (Throughput) at Constant Ψ (Purge to Feed Ratio)



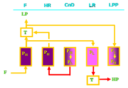
Heavy and Dual Reflux PSA Cycles

Why is the HR PSA concept important?

- CO_2 is the heavy component in flue gas and must be enriched to greater than 95 vol%
- just like in distillation, heavy component enriched with heavy reflux and light component enriched with light reflux
- nearly all PSA cycles designed to purify only the light component using light reflux
- understanding of heavy reflux PSA cycles lacking and sorely needed

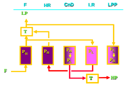
Maximum Performance Based on CO_2 Purity

5-Bed 5-Step with LR and HR from CnD



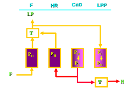
Maximum
$y_{CO2,HP}$ = 98.7 vol%
R_{CO2} = 98.7 %
(θ = 5.8 L STP/hr/kg)

5-Bed 5-Step with LR and HR from LR Purge



Maximum
$y_{CO2,HP}$ = 96.6 vol%
R_{CO2} = 71.1 %
(θ = 57.6 L STP/hr/kg)

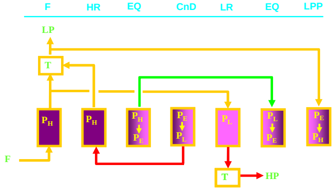
4-Bed 4-Step with HR from CnD



Maximum
$y_{CO2,HP}$ = 99.2 vol%
R_{CO2} = 15.2 %
(θ = 57.6 L STP/hr/kg)

Stripping PSA Cycle with HR and EQ

Fraction of CnD Effluent Used as HR with Remaining CnD Effluent Taken as Heavy Product



Simplest HR cycle, with one equalization step between beds 3 and 6.

Unequal Step-Time Scheduling for PSA Cycles

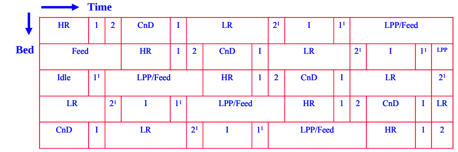
Why is PSA cycle scheduling important?

- feed step time can be lengthened → increases feed throughput
- pressure changing step times can be shortened → increases feed throughput or productivity
- feed can be delivered simultaneously to multiple beds
- multiple configurations possible, depending on the number of beds, and the number and types of steps

5-Bed 5-Step Stripping PSA Cycle with LR and HR from CnD

Two Equalization steps

Feed → HR → 1 → 2→ CnD → LR → 2[1]→ 1[1] → LPP



5-Bed 5-Step Stripping PSA Cycles with LR and HR from CnD: Effect of Equalization

Q_F = 5 SLPM, γ = 0.02, t_{cyc} = 500 sec, P_H = 138 kPa

Cycle	Number of EQ Steps	Throughput (L STP/hr/kg)	Effective Pressure Ratio	Purity (%)	Recovery (%)
I	0	θ = 57.6	12	98.0	48.5
II	1	1.5θ	6.5	97.0	36.1
III	1	1.75θ	6.5	97.2	32.1
IV	2	θ	4.7	96.8	48.0
V	2	2θ	4.7	98.5	7.5